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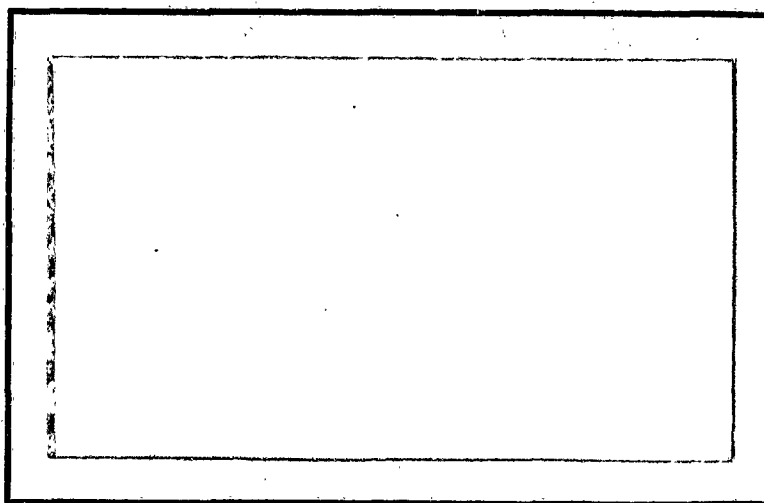
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WOODS HOLE OCEANOGRAPHIC INSTITUTION

Woods Hole, Massachusetts

Reference No. 52-14

SEA-SALT NUCLEI STUDIES

conducted during the period

October 1, 1951 - December 31, 1951

The Contractor shall furnish the necessary personnel and facilities for and, in accordance with any instructions issued by the Scientific Officer or his authorized representative, shall conduct research on the practical aspects of atmospheric sea-salt nuclei, which shall include, but not necessarily be limited to, the following:

(1) role of atmospheric sea-salt nuclei in the formation of rain with emphasis upon the relationship between weight and number of nuclei and the occurrence of rain;

(2) distribution of airborne sea-salt as a factor in the corrosion of structual steels, the primary investigation to determine how far inland sea-salt is carried by the winds, and the rates at which it is deposited on exposed structures; and

(3) determination of the water vapor pressures over concentrated sea water at lower temperature, and the study of super saturation of droplets of concentrated sea water.

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PART I

AIRBORNE SALT PARTICLES AND RAINFALL AT OAHU, T. H.
(A. H. Woodcock)

During this period, D. C. Blanchard and A. H. Woodcock
traveled to Honolulu in order to participate in the cloud
physics project of the Pineapple Research Institute, Hawaiian
Sugar Planters' Association, Territorial Cattlemen's Council,
and the U. S. Weather Bureau. They expect to remain in
Hawaii until mid-July 1952.

During this trip to Hawaii the tentative primary goals
are these:

1. To collect further data concerning the relationship
    between the numbers of large salt particles in the
    air and the occurrence of rain on Oahu (see first
    results during June 1951, WHOI Reference No. 51-87).

2. To aid local people in setting up a routine daily
    sampling of atmospheric salt with the objective of
    studying the long-term variations of the amounts of
    salt and of rainfall.

3. To make specific tests of the general relationship
    between atmospheric salt and rain drops which was
    proposed in WHOI Reference No. 51-71. These tests
    will involve sampling airborne salt particles and
    at the same time and in the same area determining
    the size and chlorinity of rain drops at the base of
    rain-producing clouds.

4. To take part in seeding experiments in which water,
    sea water, salt or other finely divided particles
    will be introduced into warm trade wind cumulus
    clouds.

The primary emphasis in the work so far accomplished has
been in the first category listed above. Forty-two samples
of atmospheric salt have been taken at cloud levels on the
windward side of the island of Oahu. These samples were taken
from an airplane over the open sea and during a period of
fourteen days. This series of measurements, when combined
with the data obtained during twelve days last June, results
in a total of twenty-six days observations of atmospheric
salt. These observations are now being analyzed and will
then be related to rainfall measured on Oahu during the samp-
ling days.

Concerning category three above, numerous rain water and rain drop samples have been taken within rain-producing cumulus clouds*. These samples were taken during a three-day period at the 3,000-foot level on the windward slope of Mt. Haleakala on the island of Maui. Preliminary examination of the drop size distribution and the chlorinity of these rain samples gives evidence in support of the hypothesis that the larger sea-salt particles become rain drops. These results will be analyzed further and presented in the next quarterly report.

In the near future simultaneous measurements will be made of the size of salt particles in the clear air at cloud levels, and the chlorinity of rain waters, and the size of rain drops. As before, the rain waters will be sampled within the rain-producing clouds on a moutain slope. These simultaneous observations should result in a clearer test of the role of sea-salt nuclei in the formation of rain.

Work is expected to begin in categories two and four within the next quarter.

PART II

PRECIPITATION STUDIES (D. C. Blanchard)

A. The Modification of the Cl Content of Rain Water by Evaporation, Coalescence with Airborne Salt in the Sub-Cloud Layer and by a "Washing Out" Action.

A proposed relationship between atmospheric sea-salt and rain drops has been advanced by Woodcock (7). It is shown that large airborne salt particles may well be the nuclei around which rain drops form, first by condensation and later by accretion. This hypothesis necessitates assuming that a direct relationship exists at the cloud base between the instantaneous intensity and the salt content of a rainfall. As it has proved to be extremely difficult to reach the cloud base, most of the attempts made to date to verify this relationship have consisted of obtaining the necessary measurements at the ground level. However, this gain in the ease of obtaining data may, and undoubtedly often does, introduce errors which

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* Sampling rains in clouds is necessary because rain drops which have fallen hundreds of meters from cloud base to earth may be modified by evaporation or by collecting salt particles by accretion. These effects are discussed in some detail by Mr. Blanchard in Part II of this report.

render the data invalid. Evaporation of the rain drops as they fall in regions of unsaturated air and pickup of salt by accretion in the sub-cloud air can both give an increase to the salt or Cl concentration of the rain water. Furthermore, evaporation not only gives an apparent increase in Cl concentration, but also lowers the intensity of rainfall. The effects of evaporation and accretion in the sub-cloud layer have been studied and their effect on the Cl versus rain intensity relationship will be discussed.

The rate of evaporation of a freely falling rain drop is a function of temperature, relative humidity, and drop diameter. Kinzer and Gunn (1) have experimentally measured the rates of evaporation of falling drops of sizes which cover the entire rain drop "spectrum". The writer has plotted on log-log paper values of dm/dt against drop diameter for an ambient temperature of 20°C. and a relative humidity of 85, 90, and 95 per cent, respectively. Between the values of drop diameter of 0.01 and 0.15 cm., the evaporation, dm/dt, may be represented as a power of the drop diameter. For the values of relative humidity mentioned above, the evaporation may be expressed by

(1) R.H. = 85; $dm/dt = -26.3 \times 10^{-6} D^{1.475}$

(2) R.H. = 90; $dm/dt = -12.84 \times 10^{-6} D^{1.475}$

(3) R.H. = 95; $dm/dt = -5.75 \times 10^{-6} D^{1.475}$

Now, as $dm/dt = \dfrac{\pi}{2} D^2\, dD/dt$, (1) becomes

(4) $D^{0.525}\, dD = -16.73 \times 10^{-6}\, dt$

By integrating from $t = 0$, $D_1 = 0.15$ cm, one obtains

an expression for the time t taken to evaporate to a

diameter of D_2 cm.

(5) $t = 2172 - 39,200\, D_2^{1.525}$

Similar expressions are easily obtainable for (2) and (3). By substituting a range of values of D_2 from 0.15 to 0.01, in steps of 0.005 cm., the time was obtained for the drop to evaporate through each size range. The maximum and minimum distance of fall of the drop in each size range was obtained by multiplying the known value of time by the terminal velocity corresponding to the initial and final drop

size, respectively. It is apparent that the actual fall distance lies somewhere between the two values and, for ease in the calculation, the average has been assumed to be the correct value. The maximum error is obtained in the size range where the terminal velocity is changing rapidly but even here, the error is only 2.2%. The distance of fall for each size range has been thus calculated for the three different cases of relative humidity. For an illustration, consider the case of relative humidity equal to 85%. Assuming a mean value of 700 meters from cloud base to ground for the general trade wind clouds of the island of Oahu, Territory of Hawaii, it is found that any drops leaving the cloud with a diameter less than 0.6 mm. will completely evaporate before reaching the ground. The larger drops are little effected. A 1.25 mm. diameter drop will reach the ground with a diameter of 1.18 mm. while a 1.5 mm. drop will arrive with a diameter of 1.44 mm.

In high intensity rains (intensities larger than 50 mm hr^{-1}) the bulk of the water is carried by drops larger than 1.5 mm. diameter. Evaporation would then be essentially negligible in modifying the intensity or Cl concentration. On the other hand, in rains of light intensity, it is often found that the bulk of the water is carried by drops considerably smaller than 1 mm. diameter. The great reduction of drop size by evaporation would have a pronounced effect on both the intensity and Cl concentration. For example, assuming that the bulk of the water in a rain of light intensity is carried by drops of 0.5 mm. diameter, one finds that they would have to fall only 500 meters in air at R.H. = 85 to evaporate to 0.25 mm. Such a reduction in diameter means a volume decrease by a factor of eight. Thus, the Cl content of the rain will rise by a factor of eight. At the same time the intensity, which is not only a function of the volume of water but also of the terminal velocity of the drops, will decrease. It will decrease by a factor of eight due to the evaporation effect and by a factor of 2.16 due to the decrease of the drop velocity (2.16 is the ratio of the terminal velocity of a 0.5 mm. drop to that of a 0.25 mm. drop). Therefore, the intensity decrease, the product of these two factors, is 2.16 x 8 or a factor of 17.3. This effect will also manifest itself at comparatively high humidities. At a relative humidity of 95 and after a fall of 700 meters, a 0.5 mm. drop will have evaporated to 0.4 mm. The volume is decreased by a factor of 1.95, the terminal velocity by 1.28 and, therefore, the intensity will decrease by a factor of 2.5. This, in turn, will increase the Cl content by a factor of 2.5.

The second mechanism which may modify the initial Cl content of a rain sample is the addition, by accretion, of airborne

salt in the sub-cloud layer. The rate of accretion depends not only on the distribution of the rain drops in the rain but also on the size distribution of the salt particles. A further complicating factor is the exponential decay rate of the airborne salt with time. For the present treatment it will be assumed that a continual supply of salt particles replenishes those lost by the accretion process. The exponential decrease of an initial concentration in a closed system will be discussed later in the report.

It is impossible to specify a drop size distribution for a given rate of rainfall. Statistical averages can and have been obtained (4) but individual samples often are far from the average. Therefore, it is not correct to compute the salt particle accretion by a given rain by merely noting the intensity of rainfall. Rather, the amount of salt picked up by any given drop falling through a known salt particle distribution must be computed. Then, if the drop size distribution in a rain is known, the salt pickup by accretion by each size range can be quickly obtained. Computations have been made on the basis of a fall height of 500 meters, assuming no evaporation. The salt particles have been assumed to be crystalline spheres with a density of 2.2. Collection efficiencies of a given drop for airborne sea-salt were made for each of six size ranges of salt particle diameter. This process was carried out for nine different drop sizes embracing the general rain drop "spectrum". Consequently, a total of fifty-four collection efficiencies were considered. Langmuir's (2) formulae were used to compute the collection efficiencies. It was necessary to compute new values as the density of sea-salt is different than that of water. The distribution of airborne salt particles was obtained from Figure 3 of Woodcock's (7) paper.

The resulting computations show that drops of 2 to 6 mm. diameter will pick up sufficient salt in a fall of 700 meters to give a Cl content of from 0.4 to 0.1 mg l^{-1}. Drops below 2 mm. in size will have a Cl content which increases with decreasing drop diameter reaching a value of 1.5 mg l^{-1} at a drop size of 0.1 mm. This increase in Cl content by accretion is small as compared to evaporation effects but, nevertheless, must be considered. The very fact that it gives a relatively greater Cl concentration for small drops than for large and, therefore, for low intensity than for high intensity rains warrants attention.

In the foregoing treatment it was assumed that the salt content of the air was constant and not subject to decrease by a washing action of the rain. It has been found, however,

that in some cases a continual decrease in airborne particles
will be concurrent with a prolonged rain (5). An idea of the
rate of "washing out" of the atmospheric sea-salt may be obtained
by reference to the usual mathematical methods used in dealing
with decay rate problems.

Let: S = g salt per cc air at time t

$S_0 = 10^{-11}$ g cm^{-3} (g salt per cc of air, initial
concentration)

$N = 0.2$ sec^{-1} (number of drops passing unit area
per unit time)

$A = 10^{-2}$ cm^2 (cross section area of 1 mm drops)

$E = 0.5$ (an average collection efficiency)

$h = 5 \times 10^4$ cm (height from cloud base to ground)

t = time in seconds

S_w = the amount of salt picked up by the rain up
until time t

Thus:

(6) $dS = -NAES\, dt$

By integrating and evaluating the constant at the
boundary conditions $t = 0$, $S = S_0$, one obtains

(7) $S = S_0 e^{-NAEt}$

As (7) gives the amount of salt left in the air at
time t, the amount "washed out" by rain is $S_w = S_0 - S$
or

(8) $S_w = S_0 (1 - e^{-NAEt})$

This gives the rate of salt pickup by water drops in
passing through 1 cm. of air. In order to obtain the value
for the total sub-cloud layer merely multiply by h. This is
not important however. The important thing is the rate of
decrease of the exponential term. Using the values given one
finds that 40% of the salt is removed by 8 minutes and 95% by
50 minutes. With a collection efficiency of unity instead of
0.5, 63% of the salt is carried away within 8 minutes. Regard-
less of what the value of E should be, it is apparent that

considerable salt is removed within 15 minutes of the start of
a storm. If a continual supply of salt laden air was not
exposed to the rain, this exponential decrease action would be
significant. In the Manoa Valley on the westward side of the
island of Oahu, T. H., where most of the rain water samples
have been obtained, it appears that the sub-cloud air is con-
tinuously being replenished so it does not seem likely that
"washing out" effects need be considered.

B. Cl Content of Rain.

The theory of rain formation as a consequence of the
introduction of large salt particles in a cloud requires that
an inverse relationship exists between rain intensity and
chlorinity. The experimental verification of this theory
must be made by analyzing water from rains of different inten-
sities. As it is unusual in Hawaiian rains to find the inten-
sity remaining constant for more than a minute at a time,
water samples must be obtained quickly. This is being accom-
plished with the aid of a large metal cone, 0.5 m^2 in area.
Three commercial windshield wipers are so mounted on the
inside that they can be rotated over the inner surface by
means of a handle extending through a hole in a plywood cover.

Nearly a hundred samples have been obtained to date. Those
which have been analyzed indicate the expected inverse relation-
ship subject to the modifications discussed in the previous
section. The results have not, as yet, been compared with drop
size measurements so no comment will be made on that part. It
is hoped that this phase of the program may be discussed in
the next progress report.

C. Drop Size Distribution in Rain.

Nearly sixty measurements have been made of the drop size
distribution in Hawaiian rains. All measurements have been
made with the filter paper method. Nylon screens, described
in the last progress report, have been used only as an explora-
tory measure. It has been found that in the medium and light
intensity rains the filter papers provide the most satis-
factory method of obtaining drop size. Not only is the drop
imprint larger than it is on the nylon screens but the inter-
pretation of the imprint is much more quickly done. In rains
of high intensity, which are not predominant in Hawaiian rains,
filter papers tend to give splash patterns. It is in these
rains that the use of nylon screens is most advantageous.

Several of the filter papers have been examined. The number of drops m^{-3} has been determined for each 0.2 mm. size range represented in the rain. From these data a plot of the number of drops m^{-3} mm-1 spread versus drop diameter has been made. Although it is still too early to draw definite con- clusions from these plots some apparent generalizations can be made. Of thirty separate drop size measurements, only six indicate any drops larger than 2 mm. diameter and only one with drops exceeding 2.4 mm. This one sample contained drops about 3.5 mm. diameter. However, these drops did not come from the usual inversion topped trade wind clouds. They fell from a cloud which had formed on a day with no inversion. The cloud top was roughly 8,000 feet. With a sufficient up- draft and the additional cloud depth it was possible for drops to grow to large diameters.

It appears that drop sizes in semi-tropical rains from non-freezing clouds as compared to rains from middle latitude clouds have less large drops and more small ones. In general then, the total number of drops m^{-3} is greater. This is to be expected as insufficient vertical cloud structures limits the drop size, while the large numbers of airborne hygroscopic salt particles provide the nuclei for large numbers of drops.

In the near future these drop size distributions will be analyzed and compared with the distribution of airborne sea- salt.

PART III

LABORATORY STUDIES IN CONNECTION WITH AIRBORNE SALT PARTICLES
(C. H. Keith)

During the past three months, laboratory studies of sea- salt nuclei were continued to attempt to develop a theoretical basis for the study of effervescence and for the growth rates of sea-salt nuclei. The method of treating these problems and some results are outlined below.

A. Effervescence

Observation of the sea surface indicates that the most probable method for introducing sea-salt nuclei into the atmos- phere is by the ejection of droplets of salt water from a bursting bubble. This choice of method is based upon the great abundance of small bubbles at the ocean surface, due to

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the wake of foam produced by breaking of waves, and upon the
fact that direct observation of bursting bubbles shows that
several droplets are projected to heights well removed from
the surface, and that the droplets generally are of a small
enough size to be carried in the air stream.

The mechanics of the bursting bubble have been qualita-
tively discussed by Stuhlman (6), and a relation between the
heights of various drops was found by him. It was felt, how-
ever, that a more quantitative treatment was to be desired.
The method of attack is to first calculate the potential
energy of the bubble at rest at the surface of the liquid,
and then to consider that this energy is transformed during
the bursting process to the potential energy of the momen-
tarily stationary jet at its highest extremity. The calcula-
tion of the equilibrium energy consists of the determination
of the function which minimizes the integral expressing the
sum of the energy of the water raised around the bubble and
that of the extended water surface. By utilizing this func-
tion, the equations for Z_o, the height of the bubble above
the undisturbed surface, and I, the potential energy can be
determined. These are, for bubbles less than 5 mm. in dia-
meter:

$$Z_o = .906 \, r^{2.7} \, \beta^{1.7}$$

$$\beta = \left(\frac{\rho g}{\gamma}\right)^{\frac{1}{2}}$$

ρ = density, in gm/cm^3

γ = surface tension, in dynes/cm

g = 980 cm/sec.

r = radius of bubble in cm.

$$I = 4\pi r^2 \gamma + \pi \rho g \, r^4 (-.604(\beta r)^{1.692} + .028(\beta r)^{6.678})$$

Calculation of the shape of the jet by static considera-
tions was not successful, since the original jet formed does
not assume the equilibrium form, but is governed by the kine-
tic process of bursting. A treatment of this kinetic problem
may be undertaken later after experimental data has been
gathered on the height and size distribution of the drops
formed from a bubble of known radius.

An interesting discovery was made early in the quarter
and noted in the preceeding progress report; i.e., that the
introduction of a surface film increased the amount of smaller

droplets produced from a given amount of foam, as well as reducing the size of the foam patch. Later it was noted that the film must have a certain amount of surface pressure, i.e., that the surface tension of the liquid was lowered, to accomplish this effect. By examination of the first equation it will be seen that this is predicted; for example, lowering the surface tension increases β and hence Z_0, hence one might accept that the drainage of water from the top of the bubble would be speeded up, and consequently the bubble under these conditions should burst sooner. Therefore the foam patch be reduced in size. Also it can be argued from this that less coalescence of smaller bubbles takes place under these conditions, and, as indicated in Stuhlman's data, smaller droplets would be formed.

B. Growth Rates.

The knowledge of the time rate of growth of a small salt nuclei is important in determining its role in the formation of rain. Ludlam (3) performed a single calculation of this rate, using Wright's (8) equation for the vapor pressure difference between a droplet and its surrounding atmosphere. It was felt that the theory should be extended to include growth rates in humidities less than 100%.

By a development that coincides with Ludlam's the following differential equation is obtained:

$$ r \frac{dr}{dt} = \frac{k}{\rho_{H_2O}} \left(\rho_{atm} - \rho_0 + \rho_0 \frac{Q}{r^3} \right) $$

where r = radius of droplet in cm., t = time in sec.

$\frac{k}{\rho_{H_2O}}$ = diffusion constant in cm^2/sec divided by the density of water

ρ_{atm} = vapor density of the atmosphere surrounding the droplet

ρ_0 = vapor density of saturated air

Q = Wright's factor for the vapor pressure lowering = $166 \, W_{salt}$

Integrating and substituting in the following relations:

$$\rho_{atm} = \rho_0 \frac{H}{100} \quad , \text{ where } H = \% \text{ relative humidity}$$

$$r_{equilibrium} = \left[\frac{Q}{\left(1 - \frac{H}{100}\right)} \right]^{\frac{1}{3}} \quad r = f r_{eq.}$$

we get finally:

$$\frac{k}{\rho_{H_2O}} \rho_0 Q t + C = r_{eq}^5 \left(-\frac{f^2}{2} + \frac{1}{6} \ln \frac{f^2 + f + 1}{(1 - f)^2} - \sqrt{3} \tan^{-1}\left(\frac{2f + 1}{\sqrt{3}}\right) \right)$$

calculating values of the expression in brackets for various values of f, we can apply the equation to droplets of any size under any humidity conditions. Letting A = the quantity within the brackets we have for water at 20°C. and 1 atm. pressure

$$\frac{394 \mu^2/\text{sec } Q t}{r_{eq}^5} = A - A_0$$

An example will illustrate the relation, if a drop with a radius of 6μ at some low relative humidity and containing 6×10^{-10} gm of salt is plunged into an atmosphere of known higher humidity, the time that it takes to grow to 99% of the equilibrium radius should be:

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90% R. H.    4.3 sec.
95% R. H.   13.5 sec.
98% R. H.   33   sec.
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These values are exceedingly low in comparison to experimental values, but this probably is due to ignoring of heat effects and curvature effects on the vapor pressure of the droplet.

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